

JC535 U.S. PTO

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PATENT APPLICATION	FIRST NAMED INVENTOR OR APPLICATION IDENTIFIER
TRANSMITTAL	BROECKER, Franz Josef
	Express Mail Label No. _____

Application Elements

Address To: Assistant Commissioner for Patents
Box Patent Application
Washington, D.C. 20231

1. / X / Fee transmittal Form
(Submit an original, and a duplicate for fee processing)
2. / X / Specification Total Pages /
(Preferred arrangement set for below)

6. / / Microfiche Computer Program (Appendix)
7. / / Nucleotide and/or Amino Acid Sequence Submission
(if applicable, all necessary)

Descriptive title of the Invention

a. / / Computer Readable Copy

Cross References to Related Application

b. / / Paper Copy (Identical to computer copy)

Statement Regarding Fed. Sponsored R & D

c. / / Statement verifying identity of above copies

Reference to Microfiche Appendix

ACCOMPANYING APPLICATIONS PARTS

Background of the Invention

8. / X / Assignment Papers (cover sheet & document(s))

Brief Summary of the Invention

9. / / 37 CFR 3.73(b) Statement / / Power of Attorney

Brief Description of the Drawings (if filed)

10. / / English Translation Document (if applicable)

Detailed Description

11. / / Information Disclosure / / Copies of IDS Citations

Claim(s)

12. / / Preliminary Amendment

Abstract of the Disclosure

13. / X / Return Receipt Postcard (MPEP 503)

3. / X / Drawing(s)(35 USC 113)(Figs.) Total Sheets /4 /
4. / X / Oath or Declaration Total Pages /4 /

- Should be specifically itemized
14. / / Small Entity / / Statement filed in prior application
Statements Status still proper and desired
15. / / Certified Copy of Priority Document(s)
(if foreign priority is claimed)

- a. / / Newly executed (original or copy)
b. / / Copy from a prior application (37 CFR 1.63(d)
(For Continuation/Divisional with Box 17 completed)
Note Box 5 below
i. / / DELETION OF INVENTOR(S)
Signed statement attached deleting
inventor(s) named in the prior application
see 37 CFR 1.63(d)(2) and 1.33(b).
5. / / Incorporation by reference (useable if Box 4b is checked)
The entire disclosure of the prior application, from which a
copy of the oath or declaration is supplied under Box 4b
is considered as being part of the disclosure of the accompanying
application and is hereby incorporated by reference therein.

16. / / Other _____

17. If a Continuing Application, check appropriate box and supply the requisite information:

/ /Continuation / /Divisional / /Continuation-in part (CIP) of prior application No. _____

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Basic Fee.....				\$ _____
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Indep. Claims: 2	-3 = _____	x \$39./\$78. = _____		
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- A check for \$690.00 for the filing fee.
- The Commissioner is hereby authorized to charge any other fee required, including the issue fee, in connection with the filing and prosecution of this application, and to the extent necessary, applicant(s) hereby petition for extension(s) of time under 37 CFR 1.136, to be charged to our Deposit Account 11-0345.

Respectfully submitted,
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5

Isothermal operation of heterogeneously catalyzed three phase reactions

This invention relates to a process and apparatus for
10 the isothermal operation of heterogeneously catalyzed reactions involving at least three phases in the form of a gaseous phase, a liquid phase and a solid phase.

The invention relates specifically to the operation of
15 such reactions where at least one reactant is liquid and one is gaseous and where the catalyst is a solid material.

The operation of such reactions is associated with
20 appreciable difficulties. The gas-liquid transfer is frequently problematical. Moreover, isothermal conditions are difficult to achieve, isothermal being used in the sense that the heat of reaction is substantially evened out by input or removal of heat,
25 so that temporal or local temperature fluctuations in the reactor are of no consequence.

Established processes are described in G. Eigenberger, Ullmann, 5th edition, vol. 4, p. 199ff. (1992) Wiley-VCH, Weinheim, Berlin, New York.

30

EP-B 0 305 203 (US-A 4 985 230) describes the operation of heterogeneously catalyzed reactions under nonadiabatic conditions. To this end, a reactor with heat-transmitting walls is packed with monolithic catalysts. A monolithic catalyst is a coherent solid having a sufficiently large catalytic surface area that countable amounts of these bodies will suffice for

catalyzing the reaction in question to an industrially sensible extent. The monolithic catalysts have channels which are angled relative to the overall flow direction, so that the reaction fluid is routed at an acute angle from one reactor wall to the other, which is said to improve the heat transfer. The shearing stress exerted on the reaction fluid is extremely high (high pressure drop) in reactor wall vicinity and otherwise rather low (poor mass transfer). This leads to unnecessarily large pressure drops in wall vicinity. The reactor is complicated to fabricate, since the pressure drop depends decisively on the geometry between reactor wall and monolithic catalyst.

- 15 EP-B 0 201 614 (US-A 4 731 229) describes a reactor containing partly corrugated tape-form catalyst bodies whose corrugation is disposed at an inclination to the main flow axis and oppositely directed in adjacent plates, the pitch of the corrugation of the catalyst body being less than the pitch of the adjacent corrugated plates and the surface area of the catalyst body being larger than the surface area of an adjacent corrugated plate. This apparatus is not contemplated for generating gas-liquid dispersions. The complicated corrugation of the plates favors bypass formation, inhibits eddying and thus compromises mass transfer. In addition, the envisioned compact packing element does not provide for effective removal of the heat of reaction.
- 30 EP-B 0 068 862 (CA-A 1 146 148) discloses a fixed bed reactor for transfer reactions between gas phase and liquid. In this reactor, the fixed bed comprises alternating layers of plane and corrugated sheets coiled together to form a roll, the corrugated sheet comprising an open mesh material with at least an outer surface layer consisting of a high molecular weight

organic polymeric substance which will be inherently hydrophobic with respect to the liquid mentioned, and the plane sheet comprising knitted, woven or felted cloth of a textile wicking material which is 5 hydrophilic with respect to the liquid or the gas-liquid transfer reaction and which will provide an uninterrupted wicking path between the ends of the roll for the liquid mentioned. The disadvantage with this type of reactor is that the textile constituents of the 10 reactor limit the cross-sectional flow velocities. The wicking path, moreover, inhibits rapid liquid transport, thus favors the separation between gas and liquid and inhibits the mass transfer between gas and liquid. Besides, the reactor is intended for adiabatic 15 operation.

It is an object of the present invention to provide apparatus and a process for carrying out reactions involving a liquid phase, a gaseous phase and a solid 20 phase with improved mass transfer between gas phase and liquid and with isothermal processing.

We have found that this object is achieved by apparatus for carrying out reactions involving a gaseous phase, a 25 liquid phase and a solid phase, comprising

- a dispersing element for dispersing a gas phase in a liquid phase to generate a reaction fluid,
- 30 - at least one reactor which possesses an inlet, an outlet and a reactor space bounded by heat-removing walls which are spaced apart substantially uniformly along the main flow axis of the reaction fluid, and which is fitted with catalyst-coated metal fabric, and
- 35

- a feed line which routes the reaction fluid from the dispersing element to the reactor inlet and is sufficiently short that the degree of dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line.

The inventors have determined that improved mass transfer can only be obtained if the reaction fluid is a dispersion formed from the gas phase (as disperse phase) and the liquid (as dispersion medium) and the process and apparatus are designed in such a way that the dispersion, as it passes through the reactor, remains stable, ie. substantially no increase in bubble size occurs.

The reactor of the invention is designed for maintaining a high but uniform shearing stress on the reaction fluid. On the one hand, it will withstand a high cross-sectional flow velocity without attrition of the catalyst. On the other, the reaction fluid is exposed to a uniformly high shearing stress in the metal fabric. This provides for uniform mixing of the reaction fluid and hence for a constant degree of dispersion of the reaction fluid as it passes through the reactor.

The catalyst-coated metal fabric of the invention is a woven or knitted metal fabric. The wire diameter is generally in the range from 0.01 to 5.0 mm, preferably from 0.04 to 1.0 mm. The mesh size may be varied within wide limits.

These wovens or knits may be coated by the process described in EP-B 0 564 830 (CA-A 2 090 930) or EP-A 0 965 384. EP-B 0 564 830 does not expressly describe the coating of metal knits with catalyst, but

they shall be treated in the same way as woven metal fabrics. For the purposes of the present invention, knitted metal fabrics are metal fabrics formed from one continuous metal thread. Woven metal fabrics, in 5 contrast, are fabrics formed from at least two metal threads.

The coating of woven or knitted metal fabrics with catalysts may also be effected by conventional dip processes, for example according to the process 10 described in EP-A 0 056 435.

If the metal forming the woven or knitted metal fabric is itself catalytically active (possibly after a 15 treatment), coating may be dispensed with entirely.

Woven or knitted metal fabrics may be used in the form of tapes. The catalyst-coated woven and knitted metal fabrics may be corrugated by means of a toothed wheel 20 roll. The introduction of corrugated woven or knitted metal fabric in the reactor makes it possible to alter the packing density of the woven or knitted metal fabric. For instance, a plurality of layers of corrugated and smooth woven or knitted metal fabric may 25 be introduced into the reactor space. Similarly, inert metal sheets may be inserted between layers of woven and knitted metal fabric. In any event, the catalyst-coated woven or knitted metal fabrics must be introduced in such a way that the reactor space is very 30 uniformly packed between the heat-conducting walls. Uniform packing suppresses bypass formation and supports the conduction of heat to the heat-removing reactor walls, which in turn enable the reaction to be carried out under isothermal conditions.

35

In a further embodiment, the dispersing element is a liquid jet gas compressor. These conventional

dispersing means are jet pumps for conveying and compressing gases.

In jet pumps, the jet of driving liquid breaks up into individual droplets on exit from the driving nozzle. These droplets become uniformly distributed across the cross section of the mixing nozzle, entrain ambient gas by impact and friction and compress it to a higher pressure. The attainable degree of dispersion is determined by the setting of driving nozzle and diffuser. This in turn depends on the pressure of the driving liquid, the suction pressure, the counterpressure, the flow of driving liquid, the gas suction stream and the mixture stream.

15

In a further embodiment of the apparatus according to the invention, the reactor is constructed as a heat exchanger. The transmission of heat through the reactor wall is decisively increased when a fluid medium on the reactor wall surface facing away from the reactor space takes up the heat of reaction and carries it away. Such a heat exchanger reactor can be constructed as a plate type heat exchanger or as a spiral type heat exchanger. A plate type heat exchanger reactor according to the invention has an in particular square or rectangular reactor space which is subdivided by additional heat-conducting walls which force the reaction fluid to take a zigzag course through the reactor space. Where the change of direction is greatest no catalyst-coated woven or knitted metal fabric is used in order that an excessively large pressure drop may be avoided. A spiral type heat exchanger reactor according to the invention has an in particular cylindrical reactor space which is packed very uniformly with catalyst-coated woven or knitted metal fabrics. The wall spacing of the heat exchanger reactors of the invention is

preferably from 1 to 30 mm, especially from 2 to 20 mm, in particular from 4 to 10 mm.

5 The invention further provides a process for carrying out reactions involving a gaseous phase, a liquid phase and a solid phase, which comprises the steps of

- generating a reaction fluid by dispersing a gas phase in a liquid phase,
- passing the generated reaction fluid through a 10 reactor whose reactor space is fitted with woven or knitted metal fabrics coated with catalyst,
- transferring the heat of reaction at the walls which bound the reactor space, and
- separating the reaction fluid into gas phase and 15 liquid phase.

The separating of the reaction fluid may be effected using conventional separators.

20 The process is preferably carried out with the overall direction of flow of reaction fluid in the reactor being upward.

25 A further embodiment of the process according to the invention is operated with separate partial recycling of gas phase and/or liquid phase. By separate partial recycling is meant that the reaction product is separated from the gas phase and/or from the liquid phase. The remaining gas and the remaining liquid may be completely or partially redispersed and fed back to 30 the reactor.

In a further embodiment of the process according to the invention, the superficial liquid velocity in the reactor is from 100 to 600 m³/(m²·h), preferably from 35 150 to 300 m³/(m²·h). The superficial liquid velocity is the volume flow of the liquid fraction of the dispersion at the reaction conditions (pressure and

temperature) divided by the cross-sectional area of the reactor space perpendicularly to the main flow axis. Since, as a result of woven or knitted metal fabrics being introduced, the reactor space is not available to 5 the reaction fluid in its entirety, the actual microscopic superficial liquid velocity is correspondingly higher.

In a further embodiment of the process according to the 10 invention, the superficial gas velocity is from 0.5 to 15 cm/s, preferably from 2.5 to 10 cm/s. The superficial gas velocity is herein defined similarly to the superficial liquid velocity.

15 In a further embodiment of the process according to the invention, the reaction fluid in the reactor is under a pressure of from 0.1 to 200 bar, preferably from 1 to 100 bar, especially from 1 to 10 bar.

20 In a further embodiment of the process according to the invention, the reaction fluid in the reactor has a temperature of from 25 to 250°C, preferably from 25 to 200°C, in particular from 50 to 150°C.

25 The invention will now be more particularly described with reference to Figures 1 to 4.

30 Fig. 1 shows an apparatus for a three phase reaction with product recycling, cycle gas operation using a liquid jet gas compressor and a plate type heat exchanger reactor.

35 Fig. 2 shows an apparatus for a three phase reaction with product recycling, cycle gas operation using a liquid jet gas compressor and a spiral type heat exchanger reactor.

Fig. 3 shows a side view of the interior of a spiral type heat exchanger reactor.

5 Fig. 4 shows a side view of a spiral type heat
exchanger reactor.

Figs. 1 and 2 show an apparatus which, the metal fabric
20 supported catalyst in the reactor 1 having been
activated (for example, by reduction with H₂), is filled
10 with product liquid by using the circulating pump 21 to
pump the liquid from the separator 10 via the optional
preheater 16 and the feed line to the liquid jet gas
compressor 5 to the liquid jet gas compressor 6 and
from there to the heat exchanger reactor 1 and from it
15 via the feed line to the separator 9 back into the
separator 10. Cycle gas is withdrawn from the separator
10 via the feed line 11 and fed by means of the cycle
gas pump 13 via the feed line to the liquid jet gas
compressor 8 to the liquid jet gas compressor 6, where
20 the gas is compressed and simultaneously dispersed in
the liquid to form the reaction mixture. A sufficiently
short feed line to reactor 7 such that the degree of
dispersion of the reaction fluid does not substantially
alter over this distance is used to feed the reaction
25 fluid into the reactor 1. Once the circulation has been
started up with product, the feed line to the liquid
jet gas compressor 4 is used to introduce reactant, and
a constant fill level system on the separator 10 is
used to withdraw a corresponding amount of product from
30 the liquid circulation via the discharge line 14. Fresh
gas to replace the reaction gas consumed is fed into
the gas circulation via the feed line to the liquid jet
gas compressor 17, with the pressure being maintained,
and off-gas is withdrawn from the gas circulation via
35 the off-gas line 12. In the case of exothermic
reactions the heat of reaction is removed from the

reactor via the cooling circulation system 22, while in the case of endothermic reactions it is introduced.

Fig. 3 shows a side view of a spiral type heat exchanger reactor according to the invention. 31 identifies the feed for the reaction fluid into the reactor (reactor inlet). 32 identifies the reactor passage which will receive the catalyst-coated metal fabric, which will take up the entire space in more or less dense packing. 33 identifies the cooling passage, which is to receive the cooling fluid.

Fig. 4 is a side view of a spiral type heat exchanger reactor and identifies the arrangement of the feed and discharge stubs. 41: reaction fluid feed (reactor inlet), 42: cooling fluid discharge, 43: reaction fluid discharge (reactor outlet), 44: cooling fluid feed. Reaction fluid and cooling fluid are here arranged in countercurrent in order that the heat transfer may be maximized. If the amount of heat released at the reactor inlet specifically is critical with regard to, for example, selectivity and catalyst stability, then a cocurrent arrangement is advisable.

The example hereinbelow illustrates the invention.

Example

The hydrogenation of benzene to cyclohexane has an exotherm of $\Delta H = -214 \text{ kJ/mol}$.

The benzene hydrogenation product equilibrates to between cyclohexane and methylcyclopentane, unless the heat of reaction is removed and a relatively low temperature is maintained.

Studies have also shown that the reaction is substrate-limited in that the low solubility of hydrogen in benzene and cyclohexane causes the reaction mixture to

deplete in dissolved H₂ along the catalyst layer. It is therefore advantageous to use the invention to improve the supply of dissolved hydrogen.

The benzene hydrogenation process is carried out using
5 an inventive apparatus as per Fig. 2, comprising a spiral type heat exchanger reactor as per Figures 3 and 4. To this end, the reactor passage 5 mm in width,
10 25 mm in depth and 960 mm in length (volume 120 ml) was packed with 8 plies of knitted catalyst fabric tape prepared by first heat-treating a knitted support tape of V2A stainless steel (German material number 1.4301) at 650°C for 3 h and then vacuum-coating it with 6 nm of platinum. The amount of active component was 46 mg.
15 The catalyst-packed heat exchanger reactor was installed in the apparatus depicted in Fig. 3. After purging with nitrogen and reduction of the catalyst with hydrogen at 80°C for 2 h, benzene was pumped via the feed line 4 into the cyclohexane-filled liquid circulation system. The reaction parameters were
20 p = 20 bar, T = 90°C and a superficial liquid and hydrogen velocity of 400 m³/m²h.

The temperature of the reaction product was measured at the reactor outlet. A maximum temperature difference of 0.2°C was observed relative to the reaction temperature
25 setting.

A selectivity of 100% was obtained with 98% conversion. The space-time yield based on the volume of the reactor passage was 0.5 kg/(l·h).

BASF Aktiengesellschaft

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We claim:-

5

1. Apparatus for carrying out reactions involving a gaseous phase, a liquid phase and a solid phase, comprising

10

- a dispersing element (6) for dispersing a gas phase in a liquid phase to generate a reaction fluid,

15

- at least one reactor (1) which possesses an inlet (31, 41), an outlet (43) and a reactor space bounded by heat-removing walls which are spaced apart substantially uniformly along the main flow axis of the reaction fluid, and which is fitted with catalyst-coated metal fabric (20, 32), and

20

- a feed line (7) which routes the reaction fluid from the dispersing element (6) to the reactor inlet (31, 41) and is sufficiently short that the degree of dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line.

25

2. Apparatus as claimed in claim 1, wherein the metal fabric (20, 32) is woven metal fabric.

30

3. Apparatus as claimed in claim 1, wherein the metal fabric (20, 32) is knitted metal fabric.

35

4. Apparatus as claimed in claim 1, wherein the dispersing element (6) is a liquid jet gas compressor.

5. Apparatus as claimed in claim 1, wherein the reactor (1) is constructed as a heat exchanger.
- 5 6. Apparatus as claimed in claim 5, wherein the reactor (1) is constructed as a plate type heat exchanger.
- 10 7. Apparatus as claimed in claim 5, wherein the reactor (1) is constructed as a spiral type heat exchanger.
- 15 8. Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 1 to 30 mm apart.
- 15 9. Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 2 to 20 mm apart.
- 20 10. Apparatus as claimed in claim 5, wherein the walls in the reactor are spaced from 4 to 10 mm apart.
- 20 11. A process for carrying out reactions involving a gaseous phase, a liquid phase and a solid phase, which comprises the steps of
- 25 - generating a reaction fluid by dispersing a gas phase in a liquid phase,
- 30 - passing the generated reaction fluid through a reactor whose reactor space is equipped with woven or knitted metal fabrics coated with catalyst,
- 35 - transferring the heat of reaction at the walls which bound the reactor space, and
- separating the reaction fluid into gas phase and liquid phase.

12. A process as claimed in claim 11, operated with separate partial recycling of gas phase and/or liquid phase.

5

13. A process as claimed in claim 11, wherein the superficial liquid velocity in the reactor is from 100 to 600 $\text{m}^3/(\text{m}^2 \cdot \text{h})$.

1

14. A process as claimed in claim 11, wherein the superficial gas velocity is from 0.5 to 15 cm/s.

15

15. A process as claimed in claim 11, wherein the reaction fluid in the reactor is under a pressure of from 0.1 to 200 bar.

20

16. A process as claimed in claim 11, wherein the reaction fluid in the reactor has a temperature of from 25 to 250°C.

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July 28, 2000

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Abstract

The invention relates to a process and apparatus for the isothermal operation of heterogeneously catalyzed reactions involving at least three phases in the form of a gaseous phase, a liquid phase and a solid phase. The invention provides apparatus for carrying out reactions involving a gaseous phase, a liquid phase and a solid phase, comprising (i) a dispersing element for dispersing a gas phase in a liquid phase to generate a reaction fluid, (ii) at least one reactor which possesses an inlet, an outlet and a reactor space bounded by heat-removing walls which are spaced apart substantially uniformly along the main flow axis of the reaction fluid, and which is fitted with catalyst-coated metal fabric, and (iii) a feed line which routes the reaction fluid from the dispersing element to the reactor inlet and is sufficiently short that the degree of dispersion of the reaction fluid does not substantially change in the course of the passage through the feed line.

Dokument-Nr.: 0227131

FIG.1

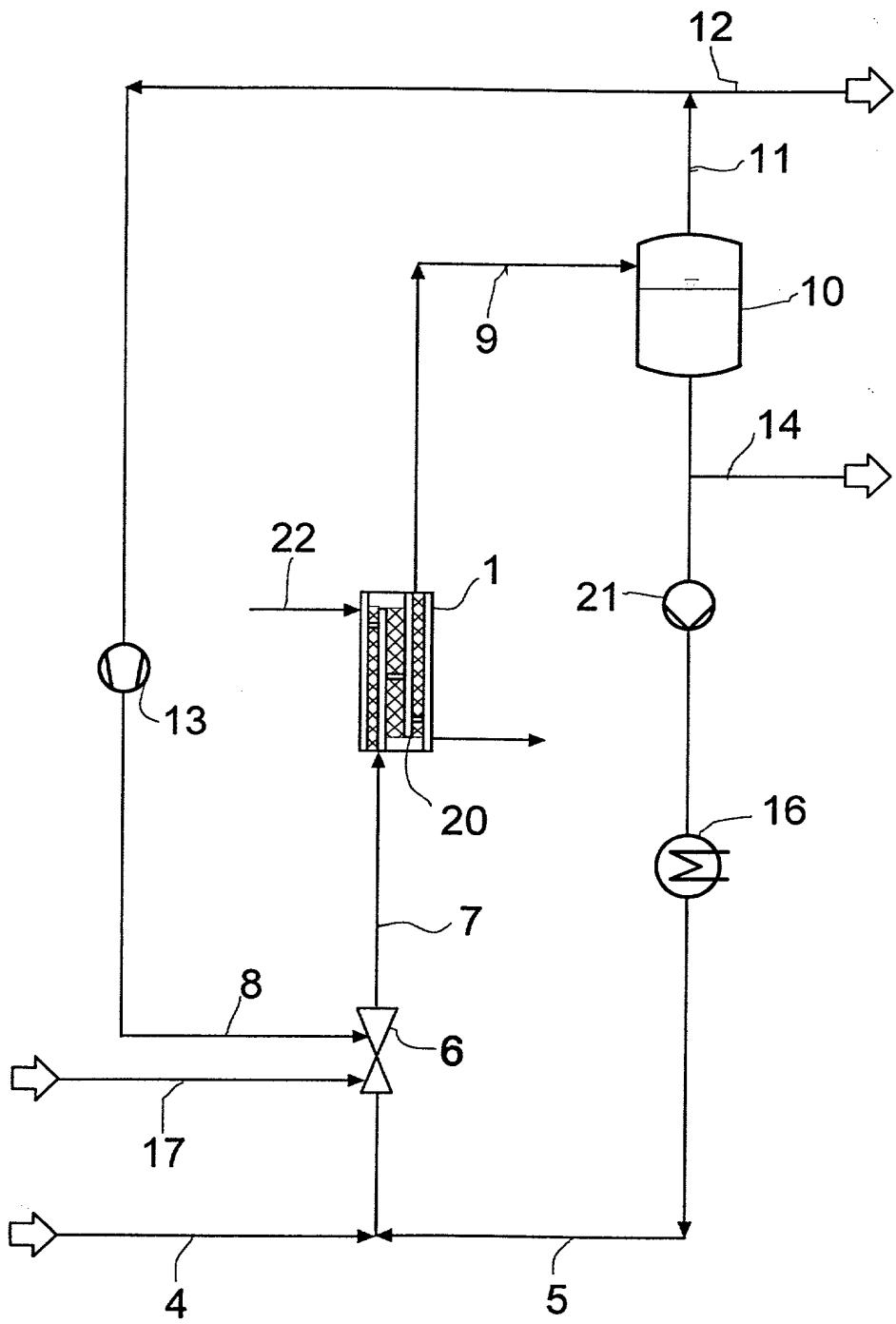


FIG.2

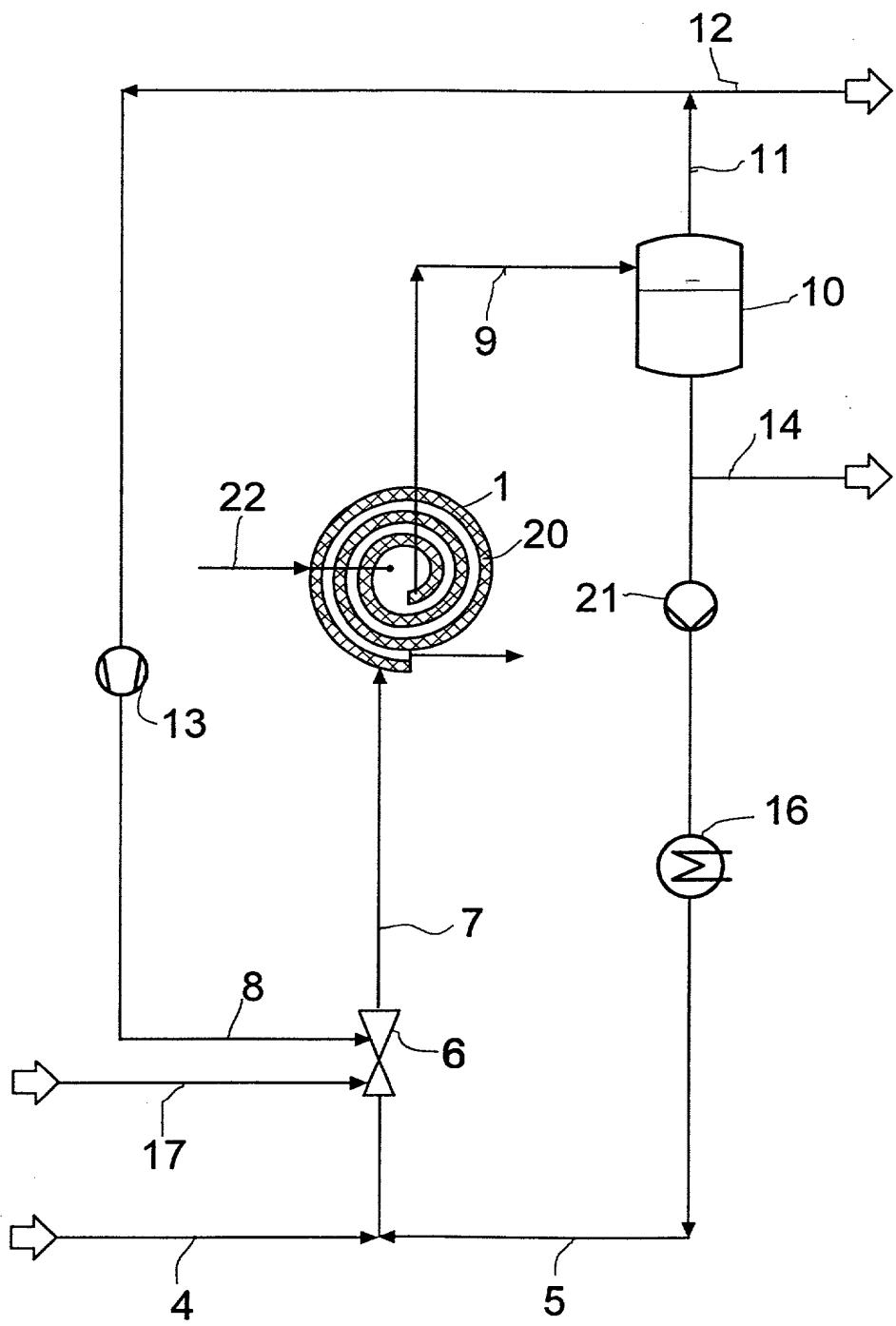


FIG.3

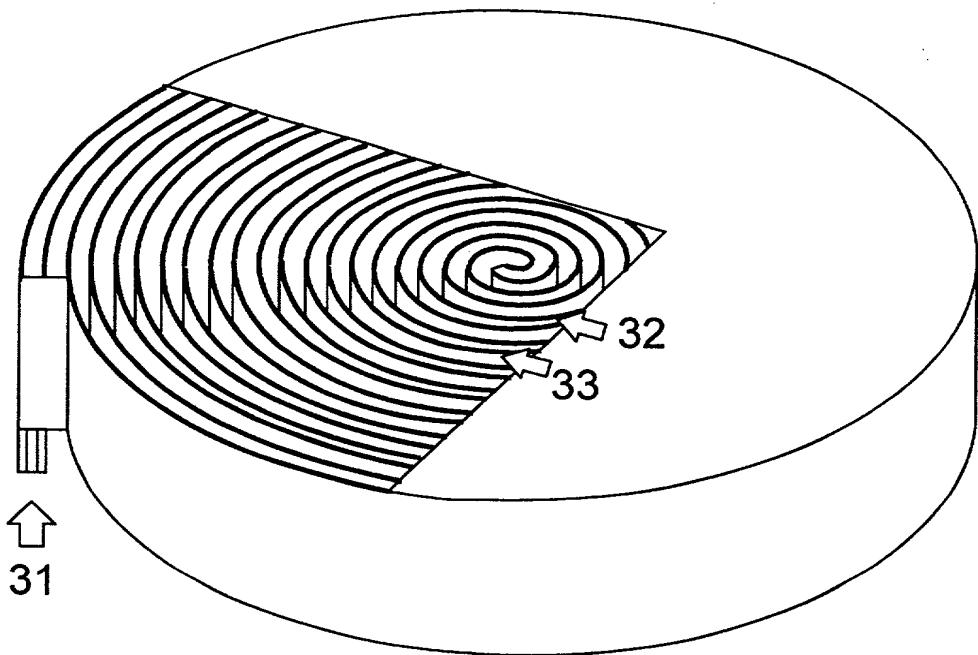
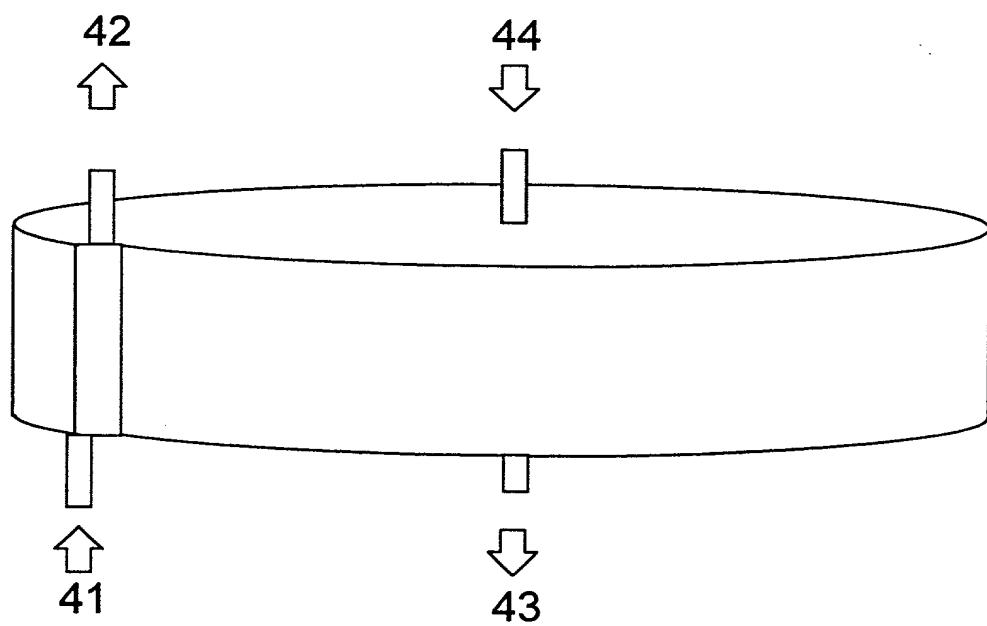


FIG.4



Declaration, Power of Attorney

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We (I), the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Isothermal operation of heterogeneously catalyzed three phase reactions

the specification of which

[x] is attached hereto.

[] was filed on 02/08/2000 as

Application Serial No. 199 36 276.9

and amended on _____.

[] was filed as PCT international application

Number _____

on _____

and was amended under PCT Article 19

on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
19936276.9	Germany	02 August 1999	[x] Yes [] No

ptskdec01-29

Declaration

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We (I) hereby claim the benefit under Title 35, United States Codes, § 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.

Filing Date

**Status (pending, patented,
abandoned)**

And we (I) hereby appoint **Messrs. HERBERT. B. KEIL**, Registration Number 18,967; and **RUSSEL E. WEINKAUF**, Registration Number 18,495; the address of both being Messrs. Keil & Weinkauf, 1101 Connecticut Ave., N.W., Washington, D.C. 20036 (telephone 202-659-0100), our attorneys, with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the patent, and to transact all business in the Patent Office connected therewith.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Declaration

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